Supplementary material belonging to:

An Experimental Study on the Kinetics of the Reaction of Fe^{II}(EDTA) with Oxygen in Aqueous Solutions

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Contents:

Absorption profiles

A typical absorption profile for a decreasing pressure experiment using N_2O is given below.

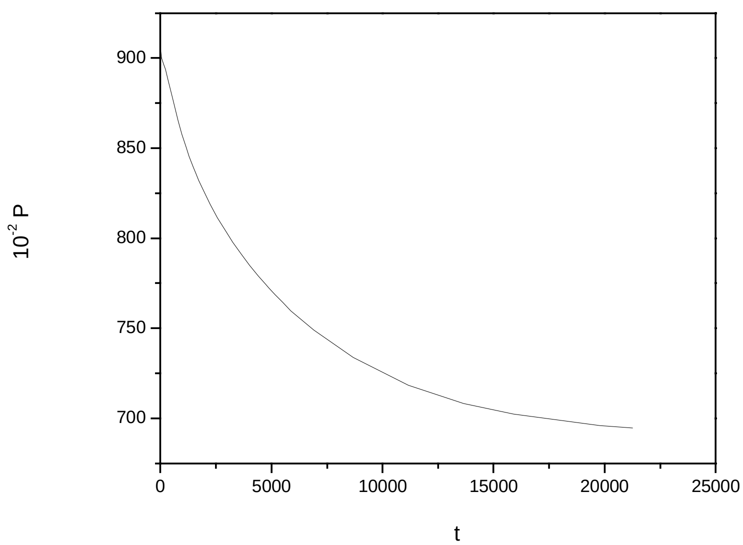

Figure 1. Absorption profile of N_2O in an aqueous Fe^{II}(EDTA) solution. T = 328 K, $C_{FeII(EDTA)}$= 60 mol/m^3, P^0_{N2O} = 90580 Pa.

2. Determination of the effect of salts on the solubility of gases

The effect of salts on the solubility of gases may be estimated using the following relation:

$$\log\left(\frac{He_{salt}}{He_w}\right) = \sum \left(h_i + h_G\right) \cdot C_i^{ion} \tag{1}$$

where h_i is a ion specific parameter, h_G a gas specific parameter and C_i^{ion} is the concentration of ions [1]. This equation can be used in the temperature range 273 - 363 K with the correction of the gas specific parameter in the following way:

$$h_G = h_{G,0} + h_T (T - 298.15) \tag{2}$$

where $h_{G,0}$ is the reference parameter and h_T is the gas specific parameter for the temperature effect.

Combining eq. (1) for O_2 and N_2O allows the elimination of the unknown values of h_i [2]:

$$\begin{gathered} \log\left(\frac{He_{O_2}}{He_{O_2\,w}} \frac{He_{N_2O_w}}{He_{N_2O}}\right) = \\ \left[\left(h_{G,0,O_2} - h_{G,0,N_2O}\right) + \left(h_{T,O_2} - h_{T,N_2O}\right)\left(T - 298.15K\right)\right] \sum_i^{N_i} C_i^{ion} = \log A_G \end{gathered} \tag{3}$$

The $h_{G,0}$ and h_T were taken from the literature [1]. For an ion concentration up to 1500 mol/m^3, A_G is 1.03 at 293 K and 1.05 at 333 K [2].

3. The effect of pH on the solubility and mass transfer coefficient of oxygen

The measured He_{O2} in Fe^{II}(EDTA) solutions at different pH's is provided in Fig. 2.

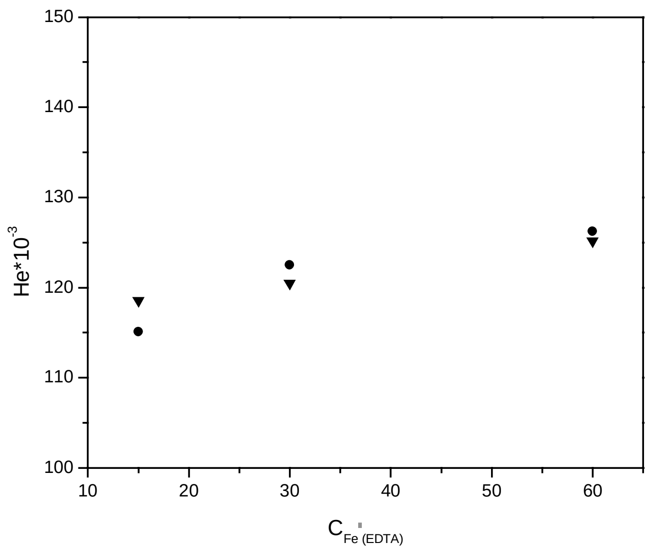

Figure 2. He_{O2} in Fe^{II}(EDTA) solutions. T = 328 K, P^0_{N2O} = 40·kPa. ●: pH = 8; ▼: pH = 5.

As expected, the He_{O2} increases with $C_{FeII(EDTA)}$. However, a clear effect of the pH on He_{O2} is absent. The effects of the pH on the k_L in Fe^{II}(EDTA) at 328 K were determined experimentally. A significant effect of the pH was absent.

4. The effect of salinity on the solubility and mass transfer coefficient of oxygen

A number of physical absorption experiments using N_2O were performed to address the effects of salinity on the k_L of oxygen ($C_{FeII(EDTA)}$ = 50 mol/m^3, T = 328 K) and the He_{O2}. He_{O2} was determined at different concentrations of dissolved salt (0 - 15 kg/m^3). The Henry coefficient was rather insensitive to the salt concentration in the concentration range applied. We therefore applied an average He_{O2} value of $2.3 \cdot 10^5$ Pa·m^3·mol^{-1} in the calculations. The k_L values were calculated and the results are given in Fig. 3. Apparently, k_L decreases when increasing the salt loading, in line with an observed reduction of the D_{O2} at higher salt concentrations [3]. In this study, the $D_{O2,w}$ decreased up to 5 % when increasing the salt concentration from 5 to 16 kg/m^3.

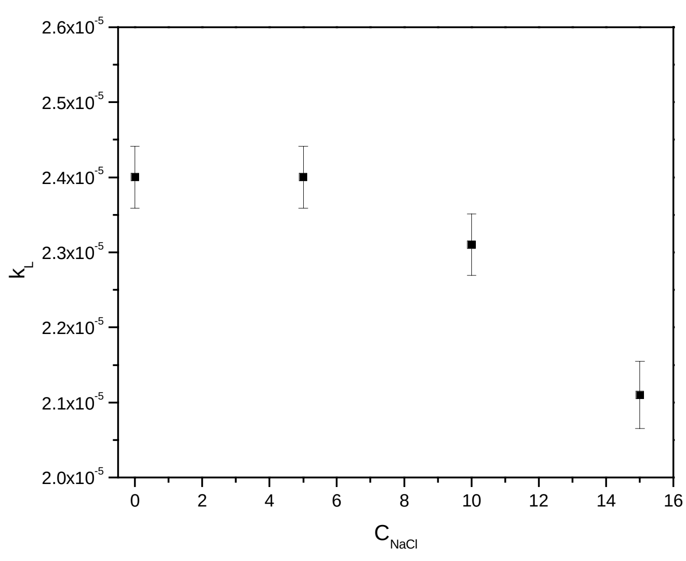

Figure 3. k_L of oxygen as a function of the concentration of C_{NaCl} at T = 328 K, $C_{FeII(EDTA)}$ = 50 mol/m^3 and pH = 7.

References for supplementary material

[1] Weisenberger, S.; Schumpe, A. Estimation of gas solubilities in salt solutions from 273 K to 363 K. *AIChE J*. **1996**, 42, 298.

[2] Demmink, J. F. *Removal of hydrogen sulfide and nitric oxide with iron chelates*. PhD Thesis, Rijksuniversiteit Groningen: The Netherlands, 2000.

[3] Van Stroe, A. J.; Janssen, L. J. J. Determination of the diffusion coefficient of oxygen in sodium chloride solutions with a transient pulse technique. *Analytica Chimica Acta* **1993**, 279, 213.